

### **REMARKS**

The Official Action dated December 23, 2005, has been carefully considered. Accordingly, the present Amendment is believed sufficient to place the present application in condition for allowance. Reconsideration is respectfully requested.

By the present amendment, claims 1 and 15 have been amended to incorporate the definition of "low elongation" set forth in the specification at page 5, lines 2-4. While it is believed that this definition was inherent in claims 1 and 15 based on the description in the specification, claims 1 and 15 now literally incorporate this language. It is believed that this change does not involve any introduction of new matter, or raise any new issue after final rejection, whereby entry is believed to be in order and is respectfully requested.

In the Official Action, the Examiner made the restriction requirement under 35 U.S.C. §121 final and withdrew claims 19-25, 28 and 29 from prosecution. As claims 19-25, 28 and 29 depend from claim 1, Applicant requests rejoinder of these claims once claim 1 is found to be allowable.

Claims 1, 2, 5-10, 12-16 and 18 were rejected under 35 U.S.C. §102(e) as being anticipated by the Gardner et al U.S. Patent Publication No. 2002/0071944. The Examiner asserted that Gardner et al disclose a breathable composite material useful as house wrap, wherein the film and nonwoven fabric layers comprise polyolefin resin compositions such as high density polyethylene (HDPE) and polypropylene. In response to Applicant's previous arguments, the Examiner asserted that the claims did not require that the low elongation material have an elongation of less than about 30% in at least one direction.

However, as will be set forth in detail below, Applicants submit that the breathable materials defined by claims 1, 2, 5-10, 12-16 and 18 are not anticipated by Gardner et al. Accordingly, this rejection is traversed and reconsideration is respectfully requested.



More particularly, as defined by claim 1, the present invention is directed to breathable material comprising a low-elongation fabric layer exhibiting less than about 30% elongation as measured according to ASTM D5034 in at least one direction, and a microporous coating thereon. The microporous coating comprises a crystalline polymer composition and a filler. As set forth at page 6 of the specification, crystalline polymer composition is defined as a polymer composition having greater than 50% of the polymer components in crystalline form. Claim 15 recites a breathable housewrap material comprising the low-elongation fabric layer and a microporous coating comprising high density polyethylene and a filler thereon. The present inventor has recognized that a crystalline polymer composition, such as, for example, high density polyethylene, in combination with the filler, will be rendered microporous by a relatively small degree of stretching which is tolerated by the low elongation fabric layer.

Gardner et al disclose a breathable composite material comprising a laminate of a nonwoven web layer and a breathable film layer. The breathability of the composite is provided by a plurality of point-like deformations of the film layer. As explained in paragraph [0015], the plurality of point-like deformations means a number of small depressions or compressed areas that penetrate into the depths of the film layer but do not perforate or form holes in the layer, and the term "point-like" is used to indicate that the deformations are essentially discrete from one another or disposed essentially continuously over a surface of the layer. Additionally, while stretching can be conducted to increase breathability, preferred composites are those in which breathability is provided by the plurality of deformations without, or with only insignificant, lengthwise or widthwise stretching (paragraph [0017]). Gardner et al disclose that heat and pressure are applied at a plurality of points on the surface of the coated web by passing the web through an embossing roll system in which at least one roll has been engraved or otherwise



machined or treated to impart a plurality of raised point or areas to the surface thereof (paragraph [0042]).

One of ordinary skill in the art will readily appreciate that the nonwoven web layer of Gardner et al is not a low elongation fabric as required by the present claims. That is, in order to effectively form the raised points or areas, one of ordinary skill in the art would appreciate that the nonwoven fabric of Gardner et al would be readily compressible and flexible. In this regard, the Examiner's attention is directed to the exemplary nonwoven materials employed by Gardner et al which are disclosed as having high elongation properties. Specifically, nonwoven Fabrics A-E of Gardner et al are disclosed as having machine direction elongations of 97%, 118%, 118%, 62% and 44%, respectively, and cross direction elongations of 188%, 252%, 252%, 166% and 78%, respectively, all of which elongations are significantly greater than the less than about 30% elongation in at least one direction required for the low elongation fabric required by the present claims. While the teachings of Gardner et al are not limited to the exemplary materials, Gardner et al provide no teaching or suggestion for use of a low elongation material, i.e., having an elongation of less than about 30% in at least one direction, as presently claimed.

Additionally, and importantly, Gardner et al broadly disclose a number of polyolefin resins for use in the film layer in paragraphs [0020], [0023] and [0033], but only exemplify a resin mixture comprising 70 parts by weight polyethylene concentrate (containing 31.7 weight % LLDPE), 10 parts by weight LLDPE and 25 parts by weight ULDPE (Example 1). One skilled in the art will recognize that low density polyethylene (LDPE) such as LLDPE and ULDPE often have low crystallinity while HDPE, a preferred material for the present microporous coating, typically has high crystallinity. In this regard, the Examiner's attention is directed to the enclosed copies of *Plastics Packaging, Properties, Processing, Applications and Regulations*, Hernandez et al, Hanser Gardner Publications, Inc., Cincinnati, OH, 2000, pp. 55-62, which discloses LDPE



as having a significantly less degree of crystallinity than HDPE (page 56), and *History and Physical Chemistry of HDPE*, pp. 1-18, [www.plasticpipe.org/drainage/pdfs/chapter201.pdf](http://www.plasticpipe.org/drainage/pdfs/chapter201.pdf), which discloses HDPE as having a greater proportion of crystalline regions than LDPE (page 4).

Applicant finds no specific teaching or suggestion by Gardner et al of a breathable material with a layer or coating formed of a crystalline polymer composition as defined in the present application, having greater than 50% of the polymer components in crystalline form, particularly in combination with a low elongation fabric.

Anticipation under 35 U.S.C. §102 requires that each and every element as set forth in the claims is found, either expressly or inherently described, in a single prior art reference, *In re Robertson*, 49 USPQ2d in 1949, 1950 (Fed. Cir. 1999). Gardner et al provide no description, express or inherent, of a breathable material as defined in claim 1 or claim 15 and comprising a low-elongation fabric layer and a microporous coating thereon, with the microporous coating comprising a crystalline polymer composition (claim 1) or high density polyethylene (claim 15), and a filler. In view of these deficiencies in the teachings of Gardner et al, Gardner et al do not anticipate the presently claimed breathable materials. Therefore, the rejection of claims 1, 2, 5-10, 12-16 and 18 has been overcome. Reconsideration is respectfully requested.

Finally, claims 3, 4 and 17 were rejected under 35 U.S.C. §103(a) as being obvious and unpatentable over Gardner et al in view of the Carroll et al U.S. Patent Publication No. 2004/0023585 or the Sheth U.S. Patent No. 4,929,303. The Examiner relied on Carroll et al and Sheth as disclosing polyethylene nonwoven open mesh fabrics.

However, as will be set forth in detail below, Applicants submit that the breathable materials defined by claims 3, 4 and 17 are nonobvious over and patentably distinguishable from the combination of Gardner et al in view of Carroll et al or Sheth. Accordingly, this rejection is traversed and reconsideration is respectfully requested.



The breathable material of claim 1, from which claims 3, 4 and 17 depend, is discussed in detail above, as are the deficiencies of Gardner et al. Neither Carroll et al nor Sheth resolve these deficiencies. That is, Carroll et al disclose a conventional nonwoven - polyolefin film laminate which is rendered breathable by incremental stretching (see Fig. 2). Applicant finds no teaching or suggestion by Carroll et al that the nonwoven is a low elongation fabric or that the polyolefin film comprises a crystalline polymer composition as presently claimed. While Carroll et al disclose that their laminate may be further bonded to a scrim material (paragraphs [0064] and [0065]), Carroll et al still fail to disclose the combination of a low elongation fabric with a film formed of a crystalline polymer composition as claimed. It is also important to note that Carroll et al only envision use of such a low elongation material after stretching of the conventional film laminate has been completed. On the other hand, according to the present invention, the claimed material may be rendered breathable by stretching the low-elongation fabric layer with the microporous coating thereon. Moreover, Applicant finds no teaching or suggestion by Carroll et al or Gardner et al for combining their teachings along the lines of the invention, or relating to any improvement provided by such a combination. Thus, the combination of Gardner et al and Carroll et al does not render the presently claimed breathable material obvious.

Sheth discloses a composite breathable film comprising a breathable polyolefin film heat laminated to a nonwoven high density polyethylene (HDPE) fabric. However, Applicant finds no teaching or suggestion by Sheth relating to the combination of a low elongation fabric with a film formed of a crystalline polymer composition, as presently claimed. To the contrary, Sheth prefers the use of a linear low density polyethylene polymer as the film layer (column 3, lines 1-26). Thus, Sheth also fails to resolve the deficiencies of Gardner et al. Moreover, Applicant finds no teaching or suggestion by Sheth or Gardner et al for combining their teachings along the lines of the invention, or relating to any improvement provided by such a combination. Thus, the



combination of Gardner et al and Sheth et al does not render the presently claimed breathable material obvious.

In the Official Action, the Examiner asserted it would have been obvious to use an open mesh as taught by Carroll et al or Sheth in the material of Gardner et al. Applicant not only disagrees, as one of ordinary skill in the art would appreciate the difficulty of making the point like deformations of Gardner et al using the scrim of Carroll et al or the nonwoven of Sheth, such a combination does not result in the breathable materials of the invention as none of the cited references teach the combination of a low elongation fabric with a film formed of a crystalline polymer composition. Further, none of the cited references teach or suggest the advantages of such a combination.

The Examiner also questioned the difference in the crystallinity of the microporous coating as claimed and the teachings of the cited references. The Examiner's attention is again directed to the references cited at pages 8-9 which describe the high crystallinity of high density polyethylene, a coating polymer composition suitable for use in the invention, and the low crystallinity of low density polyethylenes such as those exemplified in the cited references. Thus, the claimed combination of low-elongation fabric layer and a microporous coating of a crystalline polymer composition is not taught or suggested, whereby the cited combination of Gardner et al, Carroll et al and Sheth fails to render the claimed breathable materials obvious. Accordingly, the rejection under 35 U.S.C. §103 has been overcome. Reconsideration is respectfully requested.

In the event that the present application is still not in condition for allowance, entry of the present Amendment for purposes of appeal is requested.

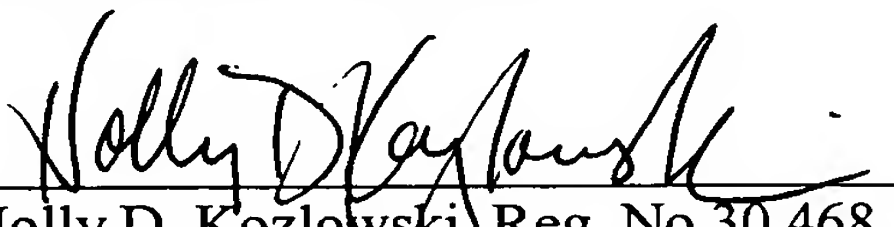
It is believed that the above represents a complete response to the rejections under 35 U.S.C. §§ 102 and/or 103, and places the present application in condition for allowance. Reconsideration and an early allowance are requested.



Serial No. 10/622,790  
Amendment Under 37 CFR 1.116 dated June 23, 2006  
Reply to Official Action dated December 23, 2005

It is believed that the above represents a complete response to the rejections under 35 U.S.C. §§ 102 and/or 103, and places the present application in condition for allowance. Reconsideration and an early allowance are requested.

Respectfully submitted,

  
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# **Plastics Packaging**


Properties, Processing, Applications,  
and Regulations

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Distributed in the USA and in Canada by  
Hanser Gardner Publications, Inc.  
6915 Valley Avenue, Cincinnati, Ohio 45244-3029, USA  
Fax: (513) 527-8950  
Phone: (513) 527-8977 or 1-800-950-8977  
Internet: <http://www.hansergardner.com>

Distributed in all other countries by  
Carl Hanser Verlag  
Postfach 86 04 20, 81631 München, Germany  
Fax: +49 (89) 98 12 64  
<http://www.hanser.de>

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Die Deutsche Bibliothek – CIP-Einheitsaufnahme

**Hernandez, Ruben J.:**

Plastics packaging : properties, processing, applications, and regulations /

Ruben J. Hernandez/Susan E. M. Selke/John D. Culter. –

Munich : Hanser; Cincinnati, Ohio : Hanser Gardner, 2000

ISBN 3-446-21404-6 (Hanser)

ISBN 1-56990-303-4 (Hanser Gardner)

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Printed and bound in Germany by Druckhaus "Thomas Müntzer", Bad Langensalza



### 3.10.1 Crystallinity

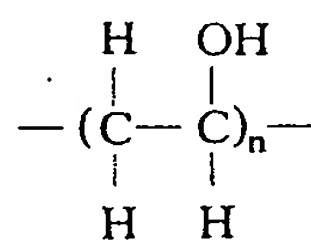
Polymer molecules tend to move towards an arrangement which is in the lowest possible energy state (lowest Gibbs free energy). The lowest energy level that a compound can achieve is a crystal form. Crystallinity involves a regular repeating arrangement of the molecules. Although crystallization tends to occur naturally, crystallization takes place in small regions of a polymer, on the order of  $1 \times 10^{-9}$  m, if at all. To produce a crystal, the polymer chains must be capable of packing closely together in a regular, parallel array. The molecules must be packed side by side in extended form, either in a planar zigzag or helical format. Polymer chains must be fairly regular in structure to permit this. Thus, the ability of a polymer to crystallize is largely determined by the regular placement of atoms in the chain, and important sources of polymer irregularity include:

1. Head-to-head or tail-to-tail placement of monomer units
2. Stereochemical irregularity
3. Copolymers with random placement of comonomers
4. Branching

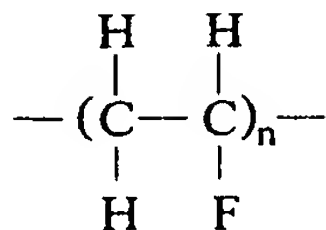
Linear polymers made of symmetrical unsaturated monomers, such as polyethylene and polyvinylidene chloride, crystallize easily. Asymmetric linear polymers such as polypropylene (PP) crystallize only if the configuration is regular, so isotactic and syndiotactic PP can crystallize, while atactic PP is amorphous.

Normally, step-reaction polymers which are synthesized from difunctional monomers containing alcohol, acid, or amines, can crystallize. These polymers produce highly ordered chains since difunctional monomers force the chain to grow in only one isomeric configuration. This is the case, for instance, with nylon 6 and nylon 6,6.

Absolute regularity is not always essential for crystallization. Some substituent groups are close enough to each other in size and behavior that they can substitute for each other randomly in a crystal lattice. Thus polyvinyl alcohol



can crystallize, even though it is atactic. Similarly, polyvinyl fluoride



is highly crystalline. In these cases, the F atom, the OH group, and H atoms are all sufficiently small that they can be "accommodated" in a crystal lattice similar to that of PE to produce highly crystalline polymers. Groups which can be accommodated within



the same crystal lattice sites, such as -H, -OH, and -F in the examples above, are called *isomorphous*.

On the other hand, polymers with regular structures and configurations do not crystallize if the substituent is excessively bulky, or if other characteristics of the polymer configuration do not permit it to assume the regular repeating pattern required. For example, when the difunctional monomers in condensation polymers contain aromatic and cyclohexane rings, only polymers with substituents in 1,4 positions, like PET and PC, are crystallizable. A 1,3 linkage in the ring causes a "kinking" of the chain which minimizes the ability to crystallize since the molecules are unable to pack in a parallel array. When a stereochemically regular polymer has a very bulky substituent group, this can interfere with crystallinity. While the presence of a bulky group does not usually prevent the formation of any crystallinity, it can significantly limit the amount of crystallinity which actually develops.

Branching also decreases crystallinity, since the branch points produce irregularities in the molecular packaging. When the reaction proceeds to cross-linking, the result is generally an amorphous polymer. Thus for step-reaction polymerization with monomers containing three and four functional groups, the resulting polymer is generally an amorphous three dimensional network. This is the case in epoxy adhesives and polyurethanes, which are amorphous thermoset materials. Low density polyethylene is able to crystallize, despite its branching, because of the very great flexibility of the ethylene chains, which contain only small hydrogen atoms as substituents. The degree of crystallinity, however, is significantly less for LDPE than for linear HDPE.

Copolymerization also introduces structural irregularity which can interfere with crystallinity. The amount of interference depends on the amount and type of comonomer, so the resultant polymer may be partially crystalline, or may be totally amorphous. When the comonomers are isomorphous, as is the case with ethylene vinyl alcohol (EVOH), a highly crystalline polymer can result even with high degrees of copolymerization. If the comonomers are not isomorphous, introducing a small amount of a comonomer will act to decrease the crystallinity of the polymer, as well as the crystalline melting temperature, whether the homopolymer associated with the comonomer is itself amorphous or highly crystalline. In most cases, random copolymers containing a substantial amount of both comonomers will be totally amorphous.

Block and graft copolymers, if they crystallize at all, tend to form small crystalline regions containing molecular segments with only one of the included monomers. Thus a block or graft copolymer may contain small regions with totally different crystalline arrangements, one type for each of the extended segments of a single monomer type.

Woodward [1] describes seven common crystalline morphologies in polymers: faceted single lamellas, non-faceted lamellas, dendritic structures, sheaf-like lamellar ribbons, spherulite arrays, fibrous structures, and epitaxial lamellar overgrowths on microfibrils. Spherulites are complex ordered aggregations of submicroscopic crystals. PE crystal spherulites, for example, are about 10 nm thick. The spherulites are separated from one another by small amorphous regions called micelles. When such spherulites are larger than the wavelength of visible light, they produce light scattering which makes



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the polymer opaque. Often plastic materials with high crystallinity are opaque, while plastics with low degrees of crystallinity are transparent or clear, and amorphous materials are totally transparent. The degree of light scattering produced by the crystallites is dependent on their size. At moderately high cooling rates, the crystalline regions can sometimes be reduced in size to less than the wavelength of light, producing a combination of high crystallinity and transparency, as is the case with some polypropylene resins and film.

We will now discuss the crystallization of polyethylene in more detail to illustrate some of the details mentioned above. A linear polyethylene molecule, HDPE, will crystallize in an extended chain conformation in the crystallite if its molecular weight is below about 10,000 daltons. Above this molecular weight, the polymer forms folded chain lamellae, as shown in Figure 3.28. The crystalline regions are closely packed chains which loop back on themselves. The region above and below the crystalline region is composed of two portions. That closest to the crystallite is a boundary region where the looping occurs. Further away is an amorphous region having no order. The molecules usually exit the crystallite after making three or four folds and enter the amorphous region. The same molecules will then either re-enter the same crystallite, or perhaps another crystallite. The morphology obtained during crystallization depends on the polymer's state of stress and on the temperature conditions.

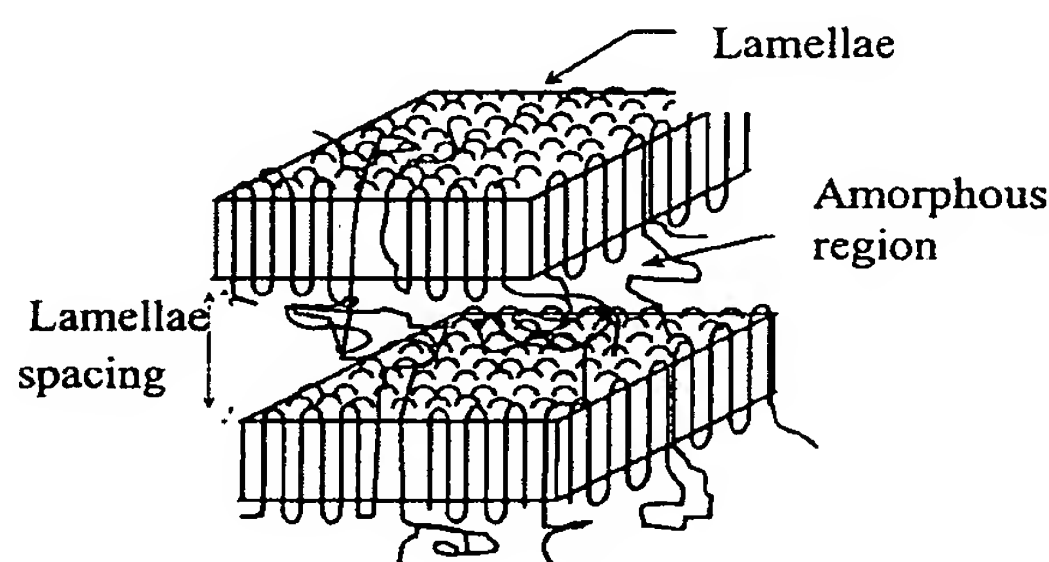


Figure 3.28 Crystallite lamellae in polyethylene.

The type of morphology obtained from a polyethylene melt differs depending on whether the melt is quiescent or under strain as crystallization occurs. In films, the melt is always under strain during the crystallization process. In an injection molded part, there will often be two regions of crystallization. The region near the walls is often under strain during crystallization, while the interior of the part may be quiescent. Another question that must be addressed about the process of crystallization to understand the probable morphology is whether or not crystallization occurred under nearly isothermal conditions. Most of the time, one can assume that nearly isothermal crystallization occurs, even in thin films. Study of non-isothermal crystallization of PE films has shown that the released heat of crystallization is enough in many cases to keep



the temperature nearly constant during the crystallization process. The critical issues are the level of strain in the melt during crystallization and the temperature gradient across the material. Non-isothermal conditions affect the amount of frozen-in strain.

In any crystallization process, the polymer molecules must slip through the melt and arrange themselves in the ordered loops of the lamellae. If there is a branch point, or a side group like vinyl acetate, these will not fit into the crystal lattice and will be rejected. The time needed to pull itself from the melt relative to the time before another molecule could begin to fit itself into the structure means that most chains develop only about four continuous folds into a crystallite before being interrupted. For polymers with regular repeating small side groups, such as polypropylene, it is possible to find a way to pack the molecules uniformly close enough together to form a crystallite. However, this requires stereoregularity. A change from the isotactic conformation of the monomer to syndiotactic would cause rejection of that portion of the molecule from the crystallite. Therefore, a regular molecular structure is very important to crystallite formation. High molecular weight and long chain branching both result in molecules having more difficulty in moving through the melt to get into the ordered structure, so both decrease crystallinity. Crystallization goes on from the time the proper temperature is reached for it to commence, until the energy levels have dropped enough that molecules no longer have enough energy to rearrange themselves into crystallites. No crystallization can occur at temperatures below the glass transition temperature or above the melt temperature (see Sec. 3.11). For many polymers, the range of temperature over which crystallization can occur is much more limited than this. As crystallization proceeds, it becomes more and more difficult to rearrange the remaining amorphous fraction of the polymer into a crystalline structure. For example, the short segments of molecules in the amorphous regions between two lamellae are constrained in their mobility by the sections which are within the crystallites. Eventually, even at relatively high energy conditions, no more crystallization can occur in the polymer. At this point, it will have both crystalline and amorphous regions. The amount of crystallinity which develops in a polymer is, therefore, a function both of its structure and of the processing conditions (energy and strain) to which it has been exposed.

In a crystallization process that occurs under strain, the morphology is described as row nucleated lamellae. Transmission electron or scanning electron microscopy show a backbone of extended chains onto which epitaxial growth of lamellae has occurred. A diagram of this structure is shown in Figure 3.29. The extended chains are higher molecular weight molecules that have been oriented in the direction of flow. The lamellae are made up of lower molecular weight species that can relax more rapidly. For this ordered growth to occur on the extended chain backbone, the chains must slip through the melt, as discussed above. Therefore, the structural characteristics that retard movement through the melt, such as high molecular weight, long chain branching, and other factors, are important influences on how fast the materials can crystallize.

Lamellae are described by an a-, b-, and c-axis. The c-axis is along the molecule. The b-axis is the growth direction, and the a-axis is the remaining direction. The stress on the melt has been shown to control the orientation of the crystallites in reference to the machine direction for films and sheet.

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High stress flows cause what is referred to as c-axis orientation along the machine direction. This would mean that in Figure 3.29, the machine direction was the vertical direction. Since one of the critical parameters for many films is the permeability, this orientation characteristic is important. Permeation cannot occur through the crystallites, so orientation in the thickness direction of the film would provide maximum barrier. This type of orientation cannot be achieved, but the right choice of polymer and processing conditions can provide a random orientation of the lamellae.

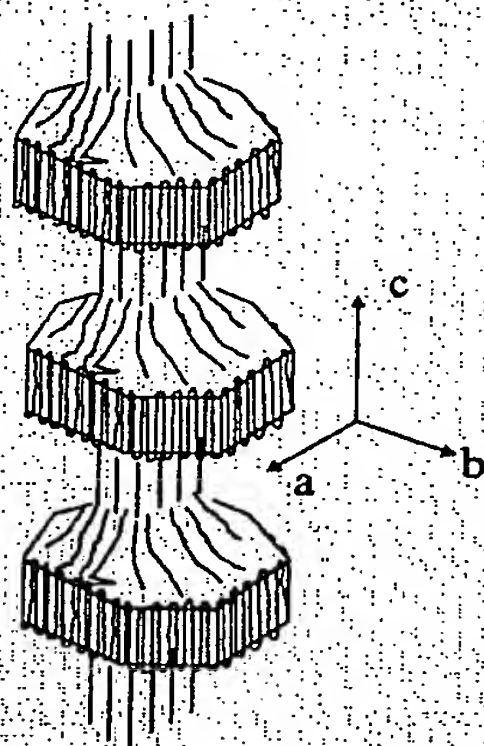


Figure 3.29 Row nucleated structure or shish-kebab, obtained from polymerization under stress.

The critical measure for characterizing a polymer is not the crystallization rate, but the elastic relaxation rate as measured by rheological means. The reason crystallization rates are not always an accurate predictor should be obvious from the preceding discussion. The rate is affected by molecular weight, molecular weight distribution, strains on the melt, and the degree and rate of cooling. Normal crystallization measurements are accomplished at a nearly quiescent state. Not all rheological measures of relaxation are necessarily accurate enough, either. Therefore, careful measurement techniques are needed to determine these parameters correctly.

As can be seen, the morphology obtained during processing can be very different depending on the conditions. The morphology that is present in the packaging material will have a pronounced effect on the properties and on the ability to further process the materials, such as material orientation, which will be discussed in the next section.



### 3.10.2 Polymer Orientation

Polymers exhibit anisotropic behavior, meaning that the properties of the material are dependent on the direction in which they are measured (as opposed to isotropic materials, where the properties do not depend on direction). Low molecular weight compounds are usually isotropic, but polymers typically show anisotropism in both the amorphous and crystalline phases. When a polymer flows, the molecules have some tendency to line up in the direction of the flow. On cooling, some of this molecular orientation is usually preserved. Therefore, stress in one direction will meet resistance from a preponderance of backbone chains with their strong covalent bonds, while stress in a perpendicular direction will be resisted by a larger percentage of much weaker intermolecular forces.

Processing conditions associated with the cooling rate of polymer melts, stretching in film blowing, and film orientation can enhance a polymer's natural anisotropy. Molecules in polymer film or sheet can be oriented into a more orderly morphology in response to external stress at temperatures above the glass transition temperature (see Sec. 3.11.2). As shown in Figure 3.30, crystallites are unraveled and reordered into fibrils. The amorphous regions are stretched also, and the increased order in the amorphous regions increases the intermolecular forces. One result is a decrease in permeability because of the effect that decreased molecular mobility has on the diffusion of the permeant through the film. The result of uniaxial molecular orientation (orientation in one direction) is a substantial increase in strength and toughness in the direction of the stretch (but a decrease in strength in the perpendicular direction). Polymers can be unoriented, uniaxially oriented, or biaxially oriented (oriented in two directions perpendicular to each other).

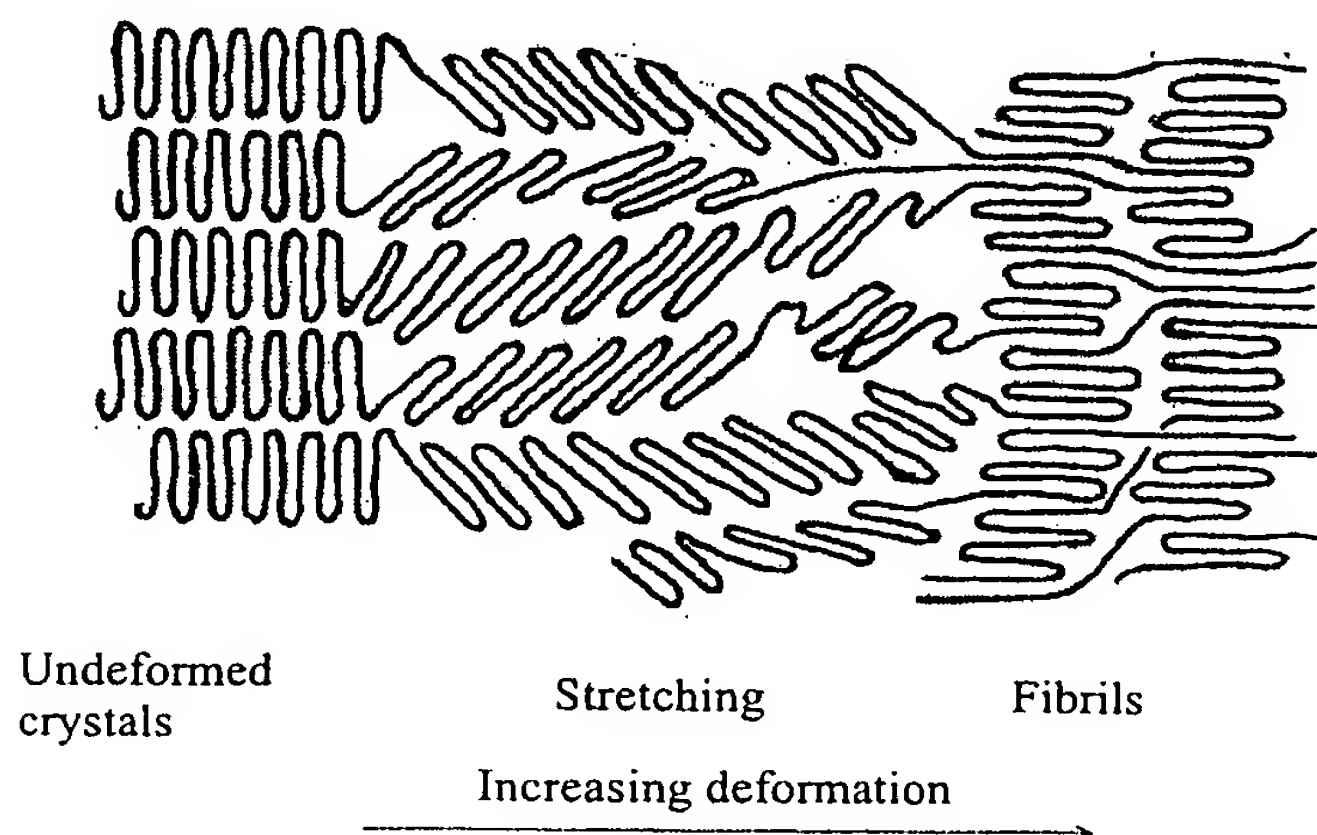


Figure 3.30 Orientation of a crystalline material, forming fibrils.

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Film used to fabricate bags is generally uniaxially oriented to improve its tensile strength, since most of the force on the bag, in use, is exerted in a vertical direction. Films used for pouches are often biaxially oriented because it is expected that tensile forces may act in both directions, and also because if shrinking occurs, the pouch should shrink in both directions.

Biaxially oriented materials can be balanced (stretched the same amount in both directions), or unbalanced (stretched more in one direction than in the other). Unbalanced oriented films will have different values of tensile and tear strength when measured in the machine direction (the direction of film travel when it is made), than when measured in the cross-machine direction (perpendicular to film travel). Unbalanced films are the most commonly produced commercially. Stretching of films is expressed as a percent of the unstretched material dimension. For example, a 400% stretch ratio indicates that the film is four times its initial length.

In PVC and PET stretch blow molded containers, molecular orientation plays an important role in the mechanical, barrier, and optical properties of the container. Orientation improves the strength and barrier capability of the container, and enhances its clarity.

At a given molecular weight and stretch ratio, the chain orientation increases with increasing strain rates and lower temperatures. Imbalances in orientation can be detected by optical techniques such as birefringence.

### 3.10.3 Degree of Crystallinity

The degree of crystallinity of a polymer reflects the relative amount of crystalline regions and of amorphous regions. This amount can be expressed on a volume or a mass basis. The degree of crystallinity is most accurately determined by x-ray scattering. In practice, this is a tedious operation and is rarely performed.

The approximate crystallinity of a polymer sample can be calculated from density measurements. Based on the two-phase model of polymer behavior, the mass or volume fraction of crystallinity can be calculated by measuring the density of a polymer sample, if the densities of the amorphous material and of pure crystals are known, as indicated by the following equations:

$$a_v = \frac{\rho - \rho_a}{\rho_c - \rho_a} \quad (3.10)$$

$$a_m = \frac{\rho_c}{\rho} a_v \quad (3.11)$$

where  $a_v$  and  $a_m$  are the volume and mass crystallinity respectively and  $\rho$ ,  $\rho_c$ , and  $\rho_a$  are the density of the sample, pure crystalline, and pure amorphous materials respectively. The percent crystallinity is  $a_v$  or  $a_m$  multiplied by 100.



The density of a polymer sample can be determined by the density gradient method, as described by ASTM D1505. In this method two solutions, A and B, are prepared with densities in the range of interest. Solution A, with the lowest density, and solution B, with the highest density, are combined in a glass tube to form a vertical column of liquid in which the density varies linearly from the bottom to the top. The column is calibrated with glass beads of known density. Plastic samples are dropped in the column and will rest at the level corresponding to their density. The density of the plastic is calculated from the position of the sample compared to that of the calibration beads.

The approximate percent crystallinity of a polymer can also be calculated from measurements of the heat of fusion (see Sec. 3.11.5) made using differential scanning calorimetry. If the heat of fusion for a pure crystalline sample of the polymer is known, the mass percent crystallinity can be determined by dividing the heat of fusion of the sample by the heat of fusion of 100% crystalline polymer, and multiplying by 100%.

A number of important polymer properties depend on the morphology of the polymer. As crystallinity increases:

Density	increases
Permeability	decreases
Opacity	increases
Blocking	decreases
Tensile strength	increases
Compression strength	increases
Clarity	decreases
Tear resistance	decreases
Impact strength	decreases
Toughness	decreases
Ductility	decreases
Ultimate elongation	decreases
Heat sealing temperature	increases
Heat sealing range	decreases

While, properly speaking, all packaging plastics are either amorphous or partially crystalline, in practice the term crystalline polymer is generally used to refer to a polymer that has any substantial degree of crystallinity. The term semicrystalline is sometimes used for polymers that have only a slight amount of crystallinity. The term paracrystalline is used to refer to materials, such as polyacrylonitrile, that have a substantial degree of molecular order, but which fall short of true crystallinity. Polymers with only a very limited ability to crystallize are sometimes, incorrectly, referred to as amorphous polymers. Polyvinyl chloride, for example, has an extremely small ability to form crystals since it is primarily atactic but has some syndiotactic tendency. Nonetheless, it is often reported to be an amorphous polymer.



**Chapter**  
**1**

**History  
and  
Physical Chemistry  
of  
HDPE**

**Lester H. Gabriel, Ph.D., P.E.**



# HISTORY AND PHYSICAL CHEMISTRY OF HDPE

## History of HDPE and HDPE Pipe

At the very close of the 19th century, German chemist Hans von Pechmann noted a precipitate while working with a form of methane in ether. In 1900, German chemists Eugen Bamberger and Friedrich Tschirner identified this compound as polymethylene, a very close cousin to polyethylene. Thirty years later, a high-density residue was created by an American chemist at E.I. du Pont de Nemours & Company, Inc., Carl Shipp Marvel, by subjecting ethylene to a large amount of pressure.

Working with ethylene at high pressures, British chemists Eric Fawcett and Reginald Gibson created a solid form of polyethylene in 1935. Its first commercial application came during World War II, when the British used it to insulate radar cables. In 1953, Karl Ziegler of the Kaiser Wilhelm Institute (renamed the Max Planck Institute) and Erhard Holzkamp invented high-density polyethylene (HDPE). The process included the use of catalysts and low pressure, which is the basis for the formulation of many varieties of polyethylene compounds. Two years later, in 1955, HDPE was produced as pipe. For his successful invention of HDPE, Ziegler was awarded the 1963 Nobel Prize for Chemistry.

Today, plastic materials used for pipes are classed under thermosetting or thermoplastic resins. Plastic highway drainage pipes belong almost entirely to the thermoplastic group (most commonly, high-density polyethylene (HDPE), PVC and ABS). They exhibit attributes of toughness, flexibility, chemical resistance and non-conducting electrical properties. Thermoplastic highway drainage pipes have been used for highway drainage since the early 1970s. Since then, growing out of applications for agricultural drainage, more HDPE drainage pipes have been installed than all other plastic pipes combined. They are being used for storm sewers, perforated underdrains, storm drains, slope drains, cross drains and culverts.

## Physical Chemistry and Mechanical Properties of HDPE

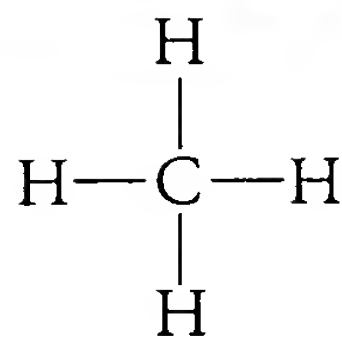
*High-density polyethylene (HDPE)* ( $0.941 \leq \text{density} < 0.965$ ) is a thermoplastic material composed of carbon and hydrogen atoms joined together forming high molecular weight products as shown in Figure 1-1c. Methane gas (Figure 1-1a) is converted into ethylene (Figure 1-1b), then, with the application of heat and pressure, into polyethylene (Figure 1-1c). The polymer chain may be 500,000 to 1,000,000 carbon units long. Short and/or long side chain molecules exist with the polymer's long main chain molecules. The longer the main chain, the greater the number of



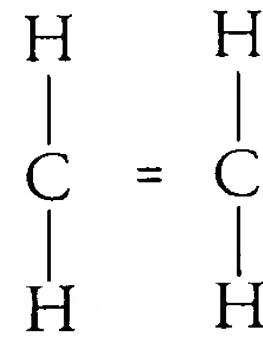
atoms, and consequently, the greater the molecular weight. The molecular weight, the molecular weight distribution and the amount of branching determine many of the mechanical and chemical properties of the end product.

Other common polyethylene (PE) materials are *medium-density polyethylene (MDPE)* ( $0.926 \leq \text{density} < 0.940$ ) used for low-pressure gas pipelines; *low-density polyethylene (LDPE)* ( $0.910 \leq \text{density} < 0.925$ ), typical for small-diameter water-distribution pipes; *Linear low-density polyethylene (LLDPE)*, which retains much of the strength of HDPE and the flexibility of LDPE, has application for drainage pipes. Less common PE materials are ultra-high molecular weight polyethylene (UHMWPE) (density  $> 0.965$ ) and very low density polyethylene (VLDPE) (density  $< 0.910$ ). Other thermoplastic materials used for drainage pipes are polyvinyl chloride (PVC), polypropylene (PP), polybutylene (PB) and acrylonitrile-butadiene-styrene (ABS).

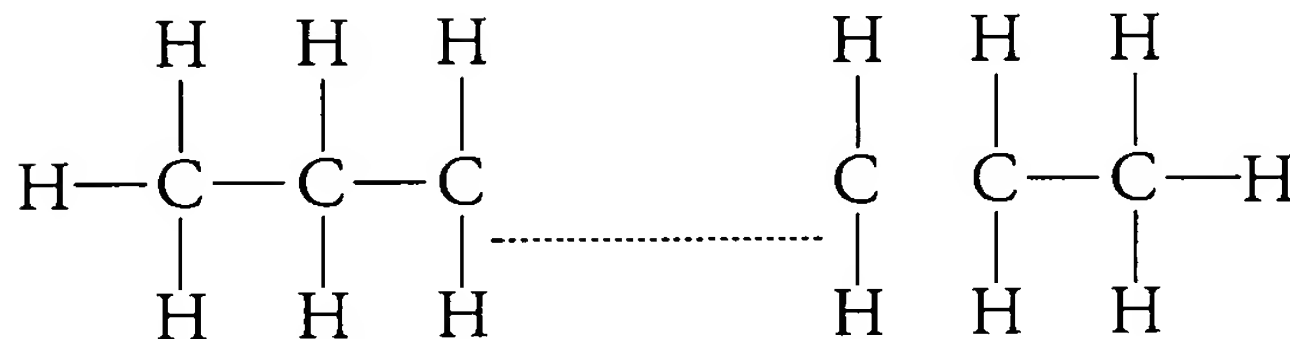
**Figure 1-1a,b,c**



**Figure 1-1a: Methane**



**Figure 1-1b: Ethylene**



**Figure 1-1c: Polyethylene Molecular Chain**

The property characteristics of polyethylene depend upon the arrangement of the molecular chains. The molecular chains, shown schematically in Figure 1-1c, are three-dimensional and lie in wavy planes. Not shown, but branching off the main chains, are side chains of varying lengths. The number, size and type of these side chains determine, in large part, the properties of density, stiffness, tensile strength, flexibility, hardness, brittleness, elongation, creep characteristics and melt viscosity that are the results of the manufacturing effort and can occur during service performance of polyethylene pipe.



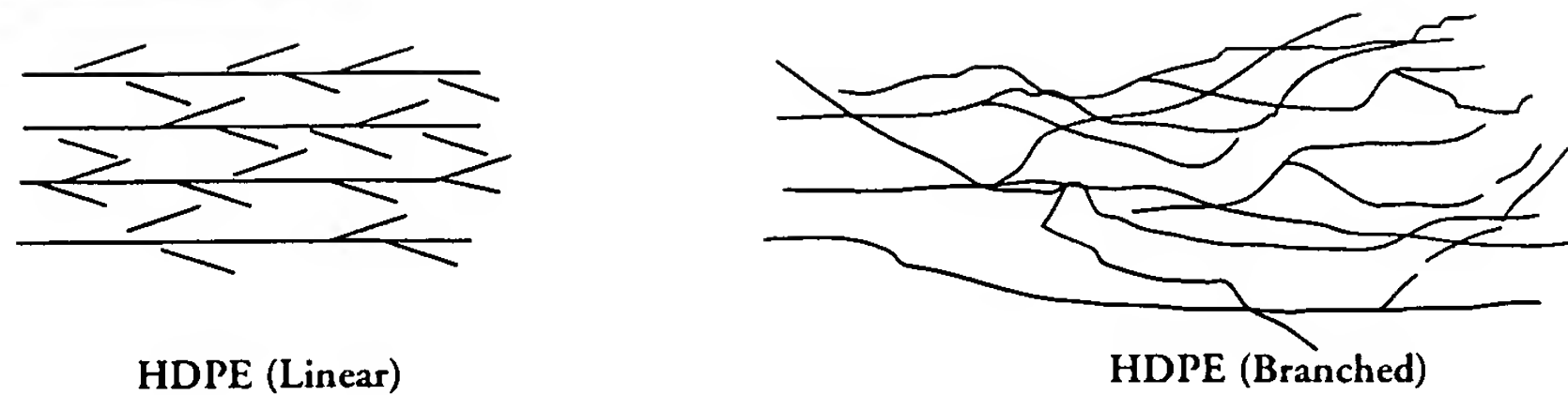
Polyethylene is characterized as a semi-crystalline polymer, made up of crystalline regions and amorphous regions. *Crystalline* regions are those of highly ordered, neatly folded, layered (in parallel) and densely packed molecular chains. These occur only when chains branching off the sides of the primary chains are small in number. Within crystalline regions, molecules have properties that are locally (within each crystal) directionally dependent. Where tangled molecular chains branching off the molecular trunk chains interfere with or inhibit the close and layered packing of the trunks, the random resulting arrangement is of lesser density, and termed *amorphous*. An abundance of closely packed polymer chains results in a tough material of moderate stiffness.

High-density polyethylene resin has a greater proportion of crystalline regions than low-density polyethylene. The size and size distribution of crystalline regions are determinants of the tensile strength and environmental stress crack resistance of the end product. HDPE, with fewer branches than MDPE or LDPE, has a greater proportion of crystals, which results in greater density and greater strength (see Figure 1-2). LDPE has a structure with both long and short molecular branches. With a lesser proportion of crystals than HDPE, it has greater flexibility but less strength. LLDPE structurally differs from LDPE in that the molecular trunk has shorter branches, which serve to inhibit the polymer chains becoming too closely packed. Hypothetically, a completely crystalline polyethylene would be too brittle to be functional and a completely amorphous polyethylene would be waxlike, much like paraffin. Upon heating, the ordered crystalline structure regresses to the disordered amorphous state; with cooling, the partially crystalline structure is recovered. This attribute permits thermal welding of polyethylene to polyethylene.

The *melting point* of polyethylene is defined as that temperature at which the plastic transitions to a completely amorphous state. In HDPE and other *thermoplastic* materials, the molecular chains are not cross-linked and such plastics will melt with the application of a sufficient amount of heat. With the application of heat, thermoplastic resins may be shaped, formed, molded or extruded. *Thermosetting* resins are composed of chemically cross-linked molecular chains, which set at the time the plastic is first formed; these resins will not melt, but rather disintegrate at a temperature lower than its melting point, when sufficient heat is added.



**Figure 1-2**



**Figure 1-2: Schematic of Linear and Branched Arrangements**

During processing, elevated temperatures and energy associated with forming and shaping the polyethylene cause random orientations of molecules within the molten material to directionally align in the extruding orifice. At room temperatures, the ordered arrangement of the layered crystalline polyethylene molecules is maintained. Tie molecules link the crystalline and amorphous regions. When the capacities of the polymer chains are overwhelmed by tension, the polymer flows (alters its shape). Tensile forces (stresses) then initiate brittle fracture, evidenced by cracking. In HDPE this may occur at very high strain rates.

Once a crack is initiated, tensile forces (stresses), which were contained prior to the event of cracking, are released. These released tensile forces (stresses) are captured by the material at the leading tips of the crack, thereby greatly increasing the intensity of the stress field and the likelihood of continued cracking at that point and all points forward. The terms *stress riser* and *stress intensity factor* are used to identify and quantify the increase in the stress field at the tips of a crack. If these regions contain and adequately respond to this increased burden, then the cracks will not propagate; if they do not, crack propagation will result. This characterizes the mechanism of *slow crack growth*. Stress risers are proportional to the measure of stress. Cracks will not propagate in a stress-free environment or where the level of stress at the tip of a crack is at a sufficiently low threshold. When the tip of a propagating crack leaves a crystal, it enters the disordered, non-layered, more loosely packed, tangled molecules of the amorphous region where the energy associated with the stress field is partially dissipated as the tangled mass of molecules adjusts in time to the sustained forces.



When polyethylene is pulled at low strain rates, in those areas where stretching has taken place, elongated rearrangement of the mass will be irreversible when molecular chains begin to slip by one another. *Ultimate tensile strength* occurs when the bonds between the molecular chains are fractured. The energy that would otherwise be stored in the system and which would otherwise be available to restore the region to its original geometry, is dissipated and unrecoverable with the event of the fracture. The new arrangement of molecules alters the stress/strain response of the remaining region. With increasing load and fewer bonds to resist, the material is less stiff and therefore takes less force to cause a unit of deformation. This phenomenon is noted on a stress-strain curve by an ever-decreasing slope as the curve bends increasingly to the right as the process continues. This is what defines *strain softening*, a characteristic of polyethylene and all materials that yield under increasing load. (The curved stress-strain curve of Figure 1-3 is an example of a strain softening material.) With sustained loads, the continuing deformation is defined as *plastic flow*. If, at some point in the deformation process the deformation is maintained, the loads and resulting internal stresses relax. This process of adjustment is called *stress relaxation*.

## **Mechanical Properties and Cell Classifications**

HDPE is a non-linear viscoelastic material with time-dependent properties. A thermoplastic pipe, serving as only one component of a pipe/soil composite structure, benefits by its attribute of stress relaxation wherein stresses (forces) are shed and transferred to the soil. Predictability of performance of a pipe in service (stress, strain and deformation responses, stability) requires knowledge of the mechanical properties of the HDPE resin and knowledge of the profile geometry.

ASTM D 3350 resin cell classifications provide the means for identification, close characterization and specification of material properties for polyethylene. Manufacturers of HDPE drainage pipes may choose higher cell classifications than the minimums required by these specifications in order to optimize competing economic and performance constraints of production, handling and service.

Density, molecular weight and molecular weight distribution dominate the resin properties that influence the manufacture of the polyethylene pipe and the subsequent performance of the pipe. Table 1-1 lists cell classification properties and the ASTM specification governing the laboratory procedure that defines and determines each. (Note that melt index (MI) is inversely related to molecular weight.) Note that cell classifications for density and molecular weight are included in Table 1-1; molecular weight distribution (MWD) is not.



**Table 1-1**

<b>CELL CLASSIFICATIONS (SEE ASTM D 3350)</b>			
<b>Property</b>	<b>ASTM Specification</b>	<b>Classification</b>	<b>Classification Requirement</b>
<b>Density</b>	ASTM D 1505 – Test Method for Density of Plastics by the Density-Gradient Technique	3	0.941-0.955 gm/cm <sup>3</sup>
<b>Melt index (MI)</b>	ASTM D 1238 – Test Method for Flow Rates of Thermoplastics by Extrusion Plastometer	3	0.4 > MI ≥ 0.15
<b>Flexural modulus (E<sub>f</sub>)</b>	ASTM D 790 – Test Method for Flexural Properties of Unreinforced and Reinforced Plastics and Electrical Insulating Materials	5	758 ≤ E <sub>f</sub> < 1103 MPa 110,000 ≤ E <sub>f</sub> < 160,000 psi
<b>Tensile strength (f<sub>t</sub>)</b>	ASTM D 638 – Test Method for Tensile Properties of Plastics	4	21 ≤ f <sub>t</sub> < 24 MPa 3000 ≤ f <sub>t</sub> < 3500 psi
<b>Slow crack growth resistance</b> Environmental stress crack resistance (ESCR)	ASTM D 1693 – Test Method for for Environmental Stress-Cracking of Ethylene Plastics	2	test condition B, 24 hr duration 50% failure (max)
<b>Hydrostatic Strength Classification</b> Hydrostatic Design Basis (HDB)	ASTM D 2837 – Obtaining Hydrostatic Design Basis for Thermoplastic Pipe Materials	0	(not pressure rated)
<b>Color (C)</b>	(not specified by ASTM)	C	2% ≤ C ≤ 5%



## **Molecular Weight Distribution**

The distribution of different sized molecules in a polyethylene polymer typically follows the bell shaped normal distribution curve described by the Gaussian probability theory. As with other populations, the bell shaped curve can reflect distributions ranging from narrow to broad. A polymer containing a broad range of chain lengths is said to have a broad molecular weight distribution (MWD). Resins with this type of distribution have good Environmental Stress Crack Resistance (ESCR), good impact resistance and good processability.

A polymer with a narrow MWD contains molecules that are nearly the same in molecular weight. It will crystallize at a faster, more uniform rate. This results in a product that will hold its shape.

Polymers can also have a bimodal shaped distribution curve which, as the name suggests, seem to depict a blend of two different polymer populations, each with its particular average and distribution. Resins having a bimodal MWD contain both very short and very long polyethylene molecules, giving the resin excellent physical properties while maintaining good processability.

MWD is dependent upon the type of process used to manufacture the particular polyethylene resin. For polymers of the same density and average molecular weight, their melt flow rates are relatively independent of MWD. Therefore, resins that have the same density and melt index (MI) can have very different molecular weight distributions. The effects of density, molecular weight and molecular weight distribution on physical properties are summarized in Table 1-2.



**Table 1-2**

<b>EFFECTS OF CHANGES IN DENSITY, MELT INDEX AND MOLECULAR WEIGHT DISTRIBUTION</b>			
<b>Property</b>	<b>As Density Increases, Property:</b>	<b>As Melt Index Increases, Property:</b>	<b>As Molecular Weight Distribution Broadens, Property:</b>
<b>Tensile Strength (At Yield)</b>	Increases	Decreases	
<b>Stiffness</b>	Increases	Decreases Slightly	Decreases Slightly
<b>Impact Strength</b>	Decreases	Decreases	Decreases
<b>Low Temperature Brittleness</b>	Increases	Increases	Decreases
<b>Abrasion Resistance</b>	Increases	Decreases	
<b>Hardness</b>	Increases	Decreases Slightly	
<b>Softening Point</b>	Increases		Increases
<b>Stress Crack Resistance</b>	Decreases	Decreases	Increases
<b>Permeability</b>	Decreases	Increases Slightly	
<b>Chemical Resistance</b>	Increases	Decreases	
<b>Melt Strength</b>		Decreases	Increases
<b>Gloss</b>	Increases	Increases	Decreases
<b>Haze</b>	Decreases	Decreases	
<b>Shrinkage</b>	Decreases	Decreases	Increases

## Density

The density of polyethylene is a measure of the proportion of crystals within its mass. Crystals, a result of the layering and close packing of polyethylene molecules, are denser than the tangled, disordered arrangement of molecules in the amorphous regions. Copolymers are often used to create and control the formation of side branches. Homopolymers, with densities of 0.960 and above, are produced without copolymers and experience very little branching. To reduce the density, butene, hexene or octene are added to make a copolymer. Butene will add branches two carbon units long; hexene, four carbon units long; and octene, six carbon units long. The greater the length of the branched carbon chains, the lower the final density. ASTM D 3350 classifies polyethylene by density as follows: *high-density polyethylene (HDPE)* ( $0.941 \leq \text{density} < 0.965$ ), *low-density polyethylene (LDPE)* ( $0.910 \leq \text{density} < 0.925$ ), *medium-density polyethylene (MDPE)* ( $0.926 \leq \text{density} <$



0.940). Less commonly employed PE materials are homopolymers (density > 0.965) and *very low density polyethylene (VLDPE)* (density < 0.910). Flexural stiffness and tensile strength increase with density; the result is increasing brittleness, and decreasing toughness and stress crack resistance.

## **Melt Index**

The melt flow rate measures the viscosity of the polyethylene resin in its molten state. It is a parameter related to the average molecular weight of the resin chains of polymer extruded through a standard size orifice under specified conditions of pressure and temperature in a ten-minute period of time. The greater the lengths of molecules, the greater the molecular weight and the greater the difficulty in extruding the resin through the standard orifice. The result: resins of greater viscosity as measured by a lower melt flow rate. When the test is conducted with pressure delivered by a standard load caused by a 47.6 lb (21.6 kg) weight at a temperature of 374°F (190°C), the resulting melt flow rate is termed the melt index (MI). The greater the viscosity, the lower the melt index value.

A lower MI (higher average molecular weight) is predictive of greater tensile strength, toughness and greater stress crack resistance. However, the lower the MI, the greater the energy required, at any extrusion temperature, to extrude polyethylene resin.

The average molecular weight, as measured by the MI, does not identify the range of chain lengths within the molecules; the molecular weight distribution (MWD) does. Polyethylene polymers of the same MI and the same density may have very different properties if the molecular weight distributions (MWD) are different. A polymer with a narrow MWD will crystallize more rapidly and with greater uniformity, resulting in less warpage and greater fidelity to the intended geometry. A polymer with broad MWD may have better stress crack resistance, impact resistance and ease of processing.

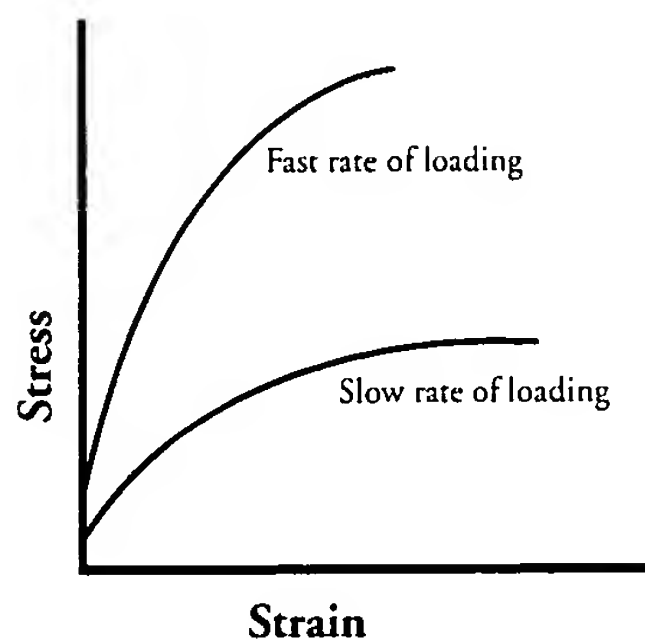
## **Flexural Modulus**

The flexural modulus ( $E_f$ ) is a material stiffness that is, in part, predictive of a structure or a structural element's resistance to bending under the application of loads. When combined with the geometric stiffness (a function of the moment of inertia and other geometric properties), the composite stiffness is termed the bending stiffness. The greater the bending stiffness, the greater the bending resistance and, other things being equal, the greater the bending stresses. For flexible pipe, the material modulus ( $E$ ) is a composite of the material's flexural stiffness ( $E_f$ ) and ring compression stiffness ( $E_c$ ). Current design practice assumes equivalence for working values  $E_f$  and  $E_c$ .



Non-linear stress/strain curves of HDPE, and the modular values derived therefrom, are sensitive to rates of load application and are generally 'linear' up to approximately 2% strain. Stress and strain are determined at the point of maximum bending on a simply supported test beam caused by a centrally applied load. The slope of the line drawn between points of zero strain and 2% strain on a stress/strain curve typically defines the flexural modulus. Because of the stress relaxation attribute of HDPE, the less rapid the loading and the longer the duration of load application, the flatter the early slope of the stress/strain curve and the lower the estimate of flexural modulus; hence the need for a carefully defined (see ASTM D 790) rate of load application. (See Figure 1-3.)

**Figure 1-3**



**Figure 1-3: Curves of Stress v Strain (immediately after loading)**

For HDPE pipes, the minimum pipe stiffness requirements set by specification determines, in part, the amount of material required, the cost of which dominates the cost of the finished pipe delivered to the job site. The characteristics of the stress/strain curve and the associated values of stress, strain and pipe stiffness are sensitive to the rates of application of load and displacement.

Stiffness requirements for pipes of any material may be met by material adjustments to the modulus of elasticity, geometric adjustments to the moment of inertia, or both. Profile pipe walls, easily shaped in HDPE by extrusion and/or vacuum forming, are designed to increase the wall's moment of inertia above that which would be the case for a solid wall pipe of the same material content, thereby enabling an optimization of cross-sectional area. The flexural modulus increases with density for a given melt index. See Table 1.2 for the effects of changes in density and melt index on the more general properties of HDPE.



## **Tensile Strength**

The point at which a stress causes a material to deform beyond its elastic region (permanent deformation) is called the tensile strength at yield. When stressed below the yield point, an elastic material recovers all the energy that went into its deformation. Recovery is possible for polyethylene when the crystals are subjected to low strain levels and maintain their integrity. A formulation of greater density (higher fraction of crystals, lower melt index) is predictive of greater tensile strength and increasing brittleness.

The force required to break the test sample is called the ultimate strength or the tensile strength at break. The strength is calculated by dividing the force (at yield or break) by the original cross-sectional area. ASTM D 638, *Standard Test Method for Tensile Properties of Plastics*, is used to determine the tensile properties of polyethylene pipe resins. Test specimens are usually shaped as a flat “dog bone”, but specimens can also be rod-shaped or tubular per ASTM D 638. During the tensile test, polyethylene, which is a ductile material, exhibits a cold drawing phenomenon once the yield strength is exceeded. The test sample develops a “neck down” region where the molecules begin to align themselves in the direction of the applied load. This strain-induced orientation causes the material to become stiffer in the axial direction while the transverse direction (90° to the axial direction) strength is lower. The stretching or elongation for materials such as polyethylene can be ten times the original gauge length of the sample (1000% elongation). Failure occurs when the molecules reach their breaking strain or when test sample defects, such as edge nicks, begin to grow and cause failure. Fibrillation, the stretching and tearing of the polymer structure, usually occurs just prior to rupture.

Tensile or compressive elastic deformations of a test specimen along a longitudinal axis excite respective inward or outward deformations parallel to a transverse axis normal to the first. Poisson's ratio is the ratio of lateral strain to longitudinal strain. When tested according to ASTM E 132, *Standard Test Method for Poisson's Ratio at Room Temperature*, Poisson's ratio for polyethylene is between 0.40 and 0.45.

## **Environmental Stress Crack Resistance (ESCR)**

Under certain conditions of temperature and stress in the presence of certain chemicals, polyethylene may begin to crack sooner than it would at the same temperature and stress in the absence of those chemicals. This phenomenon is called environmental stress cracking (ESC).



Stress cracking agents for polyethylene tend to be polar materials such as alcohols, detergents (wetting agents), halogens and aromatics. The property of a material to resist ESC is called environmental stress crack resistance, or simply ESCR. The mechanism is not fully understood, but failures from ESC tend to be due to the development of cracks in areas of tensile stress which slowly grow and propagate over time. Stress cracking may be avoided by using appropriate resin formulations of stress crack resistant materials; appropriate geometric designs and manufacturing controls that prevent the occurrence of severe stress risers; and by limiting stresses and strains during pipe installation.

There are over 40 different ESCR test methods used to determine the chemical resistance of various materials. The standard test currently used in the polyethylene industry is the bent-strip test. It is also called the “Bell Test,” since it was developed during the 1950’s for wire and cable coatings for the telephone industry. ASTM D 1693, *Standard Test Method for Environmental Stress Cracking of Ethylene Plastics*, describes the test method used to determine the ESCR value for polyethylene. Ten small compression-molded specimens are notched and bent and then placed into a holder. The holder is immersed into a tube of a surfactant, typically one such as Igepal CO-630 at 212°F (100°C) and 100% concentration, and the time to failure is noted. The results are reported using the notation Fxx, where xx is the percentage of samples that have failed. For example, the statement F20=500 hours means that 20% of the samples have failed within 0 to 500 hours.

This test was developed when the time to failure was less than 10 hours. Excellent stress crack resistance of modern resins, coupled with stress relaxation in the pre-bent samples results in a test method wherein few failures occur. The efficacy of the test diminishes after a few hundred hours. This test is currently used mainly as a quality assurance test rather than providing definitive rankings of pipe performance.

### **Notched Constant Ligament Stress (NCLS)**

Disadvantages of the ESCR test method are overcome with the Notched Constant Tensile Load (NCTL) test as described in ASTM D 5397. Because ASTM D 5397 is intended for geosynthetic materials using membranes as the specimen, a new test method was developed for piping materials – the Notched Constant Ligament Stress (NCLS) test. In this test method, HDPE resin is compression molded into a plaque. Dumbbell samples are machined from the plaque and notched in the midsection. Samples are placed in an elevated temperature bath containing a wetting agent for acceleration. The sample is then subjected to a constant ligament stress until a brittle failure occurs from slow crack growth. This is now an ASTM test method, F 2136.



## **HDPE – A Material of Choice**

Metal, plastic, concrete and clay make up most of the materials used for the manufacture of drainage pipes. Metal pipes may be steel, ductile iron or aluminum; concrete pipes may be steel-reinforced, earth-reinforced, non-reinforced, precast or cast-in-place; and plastic pipes may be of thermosetting resins (e.g., glass-reinforced epoxy or polyurethane) or thermoplastic resins (e.g., HDPE, PVC, polypropylene or ABS (acrylonitrile-butadiene-styrene)). The material longest in use is vitrified clay; the newest materials are plastic. Some pipes are built with a combination of materials; corrugated steel pipes lined and/or coated and/or paved (inverts) with plastic, bituminous or concrete materials. Durability (mostly, resistance to chemical and electro-chemical corrosion and abrasion), surety of structural performance over time, integrity of joints, surety of hydraulic performance (as pipe ages), ease of construction, availability and life cycle costs dominate the choice of pipe material(s).

Highway drainage facilities are often subject to hostile effluents and embedment soils. Concrete pipe is subject to chemical attack when in the environments of low pH (acids) and/or soluble salts (sulfates and chlorides) in drainage waters and neighboring soils. Sulfates, mainly those of sodium, calcium, potassium and magnesium, are found in many locations in the states of the northern Great Plains, in the alkali soils of western and southwestern arid regions, and in seawater. Uncoated (or otherwise unprotected) galvanized steel pipes are degraded in environments of low pH and low resistivity of soil or water. Permissible levels of pH vary by jurisdiction; a range of soil or water of  $6.0 < \text{pH} < 9.5$  is generally accepted. Unlike pipes of concrete, steel, aluminum and iron, thermoplastic and vitrified clay pipes do not corrode or otherwise degrade in these environments; expensive maintenance is not required. Unlike metal pipes and steel reinforcement of concrete pipes, thermoplastic and vitrified clay pipes are non-conductors; cathodic protection is not required to prevent degradation due to galvanic corrosion at locations of low soil resistivity or in the vicinity of stray electrical direct currents. Polyethylene is often used to line and encase metal pipes thereby offering barrier protection from aggressive soils or stray electrical currents leading to galvanic corrosion. HDPE offers a range of  $1.5 < \text{pH} < 14$ .

Accidental highway spillage of high concentrations of some organically based chemicals, such as crude oils and their derivatives (solvents, gasoline, kerosene) or concentrated acids and bases, may cause swelling and softening of thermoplastic materials if sustained over long periods (measured in months). Of the four most common drainage pipes of thermoplastic materials (ABS, PVC, polypropylene, and HDPE), resistance to these aggressive chemicals is in the order noted; ABS the least resistive, HDPE is the most resistive.



Polyester and epoxy thermosetting resin pipes, reinforced with continuous windings of glass filaments, primarily intended for sanitary sewers, were found to be corrosive in the presence of available hydrogen ion (present in acids and water). Penetration to the glass/resin interface may result in debonding of the glass reinforcement and wicking along the glass/resin interface. Thermosetting resin pipes reinforced with randomly oriented chopped fibers of short lengths have succeeded these pipes.

The chemical inertness of HDPE and the flexible “trampoline” response of the long chain molecules of HDPE result in a highly corrosion-resistant material. HDPE pipe is most often favored for transporting slurries containing highly abrasive mine tailings. Abrasion of metal, bituminous and concrete protective coatings of metal and concrete pipes (a function of the square of the flow velocity) leave these pipes vulnerable to accelerated erosion after penetration to the bare pipe material.

For the same conditions of embedment, the more flexible the pipe the lesser the proportion of overburden load attracted to the pipe. The attribute of stress relaxation of HDPE pipes (and thermoplastic pipes in general), which is greater than any relaxation of the embedding soil, assures that overburden loads and stresses within the pipe walls will decrease with time. The result is that a significant proportion of loads initially resisted by a flexible pipe will be transferred to the soil of the pipe/soil composite structure; the opposite is true for rigid pipes. Furthermore, the ability of buried flexible pipes to alter their shapes from circles to ellipses is exactly what transforms much of what would be bending stresses (which include tensile stresses) into membrane ring compression stresses. For the same conditions of embedment, rigid pipes (which lack the ability to comply with alteration of shape) respond with greater tensile stresses than flexible pipes and, in the case of concrete pipes, require steel reinforcement to manage these tensile stresses. HDPE pipes, properly embedded in a competent soil mass, result in a formidable soil/pipe composite structure that is almost entirely in the favored ring compression.

Favorable and commonly accepted roughness values of Manning’s ‘n’ of 0.010 - 0.013 make smooth-lined corrugated HDPE a favorable choice for the transport of drainage waters. Velocity of flow is insensitive to changes in pipe shape due to service loads. The non-stick surface of HDPE resists scaling and pitting, and therefore does not require a design with a less favorable Manning’s ‘n’ to accommodate future conditions.



## **Additional Considerations**

### **Crack Resistance:**

Weak molecular bonds, perpendicular to the densely packed layered molecules of polyethylene crystals, tie adjacent molecules. In response to tensile stresses, cracks may form and propagate parallel to these layers by rupturing these weak bonds. Less dense and disordered arrangements of molecules in amorphous regions are more resistant to crack propagation than the layered molecules in crystals. For polyethylene resins of the same molecular weight, the lesser the density, the greater the resistance to stress cracking. The greater the proportion of crystals, the greater the density and brittleness of the resin. Density alone, however, is an inadequate predictor of stress crack resistance.

All common materials, extruded or otherwise shaped or formed at elevated processing temperatures, shrink during cooling. Residual stresses, which result, combine with those stresses resisting externally applied loads. In processes where stretching after forming takes place result in mechanical properties parallel to the direction of stretch different than those oriented perpendicular to the direction of stretch. At low rates of strain, should cracking of these orthotropic materials occur, they are likely to be parallel to the direction of stretch.

A more general purpose of ASTM D 1693, the test for ESCR, is prediction of the performance of ethylene resins subjected to environments such as soaps, wetting agents, oils, detergents or other materials likely to be stored or marketed in containers. This test is likely to assure proper material formulation (inclusive of post-consumer recycled resins) and to minimize contaminant inclusions.

### **Compression**

The response of a buried flexible pipe is dominated by compression. Note in Table 1-1 there is no cell classification for compression. For purposes of design and for small strains (less than 2%), the compression modulus is taken to be of equal magnitude as the elastic tensile modulus. At greater stress levels, compression strain is less than the tensile strain. HDPE in compression does not tear or crack; stability for thin elements is a design consideration.



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